Local optical parameters of spherical polydispersions: simple approximations

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New analytical solutions for the local optical characteristics (extinction and absorption coefficients, asymmetry parameters of phase functions) of spherical polydispersions composed of comparatively large particles are derived. The geometric optics (GO) approximation is used to solve the problem. For the accuracy of the GO approximation to be improved, the edge effects were taken into account. A comparison with the data obtained by the use of the Mie theory shows a satisfactory accuracy of our analytical formulas. The simple formulas for the cloud local optical characteristics are derived.

Key words: Geometric optics approximation, Mie theory, edge effects, particle-size distribution, cloud optics.

1. Introduction

In this article we give analytical formulas for asymmetry parameters of phase functions, extinction and absorption coefficients for comparatively large spherical particles and their polydispersions in terms of the complex refractive index of a particle substance $m = n - i\chi$, and the particle-size distribution (PSD) $f(a)$ (a is the particle radius).

The problem of light diffraction by spheres was solved as long ago as in the 19th century.[1] We suppose that everybody who currently deals with light scattering has the Mie computation code on his or her PC.

Then what forced us to come back to this problem? Only necessity did. As is known, Mie computations become more tedious the larger the dimensionless size parameter $x = ka$ ($k = 2\pi/\lambda$, where λ is the wavelength). At the same time atmospheric aerosols, clouds, and fog contain a lot of large particles. Analytical solutions that directly relate optical and microphysical parameters of aerosols are badly needed (instead of, or along with, the Mie computation algorithms) for the biomedical community, for engineers and researchers involved in remote sensing, climate change problems, cloud optics, and so on.

A lot of attempts to simplify the calculation of the optical characteristics of spherical particles have been made.[2–6] Geometric optics (GO) partially resolves the situation, providing solutions that are much simpler than Mie formulas for particles with $x > 10^2$–10^3.[2–6] But the mean size parameters of soil and oceanic aerosols, mist drops, and cloud drops are often less than 10^2–10^3.

To improve the accuracy of the GO approximation, edge effects should be taken into account.[3,7–10] Solutions for the efficiency factors with regard to the edge effects were obtained by Nussenzveig and Wiscombe.[10] However, these solutions are also too cumbersome.

Our ultimate goal here is to give straightforward formulas for the characteristics[3] of scattering by spherical particles. These formulas are supposed to provide a sufficiently high accuracy over a wide range of x. To be more specific, we consider the following integral characteristics of scattering: scattering cross section C_{sca}, extinction cross section C_{ext}, absorption cross section C_{abs}, asymmetry parameter g for a single particle, and scattering σ_{sca}, extinction σ_{ext}, and absorption a_{abs} coefficients, as well as asymmetry parameters $\langle\cos\theta\rangle$ of phase functions for spherical polydispersions. All our formulas rely on simplifications of the known GO solutions and include additional terms (the edge terms) that allow for edge effects. To derive completely analytical solutions, we obtained the numerical coefficients of the edge terms through the approximation of Mie computations. We have successfully used these derived formulas to solve

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different applied problems, including one for cloud–radiation interaction.

2. Simplification of Geometric Optics Solutions

As is known, within the framework of the GO approximation, with allowance for diffraction, the extinction cross section $C_{\mathrm{ext}}{}^{G}$, the scattering cross section $C_{\mathrm{sca}}{}^{G}$, and the asymmetry parameter g^{G} of the phase function for a large spherical particle are given by the following relations[2,6,10]:

$$C_{\mathrm{ext}}{}^{G} = 2\pi a^2, \tag{1}$$

$$C_{\mathrm{sca}}{}^{G} = \pi a^2 (1 + Q_{\mathrm{sca}}{}^{G}), \tag{2}$$

$$g^{G} = \frac{1 + y}{1 + Q_{\mathrm{sca}}{}^{G}}, \tag{3}$$

where

$$y = \frac{1}{2}\sum_{j=1}^{2}\int_0^{\pi/2} \times \frac{\phi_j(\tau)\sin 2\tau \mathrm{d}\tau}{1 - 2R_j \exp(-c\xi)\cos 2\tau' + R_j{}^2 \exp(-2c\xi)}, \tag{4}$$

$$Q_{\mathrm{sca}}{}^{G} = \frac{1}{2}\sum_{j=1}^{2}\int_0^{\pi/2} F_j(\tau)\sin 2\tau \mathrm{d}\tau. \tag{5}$$

Here

$$\begin{aligned}\phi_j(\tau) &= \exp(-c\xi)(1 - R_j)^2 \cos 2(\tau - \tau') \\ &\quad + R_j[1 - \exp(-2c\xi)]\cos 2\tau \\ &\quad + 2R_j{}^2[\exp(-c\xi) - \cos 2\tau']\exp(-c\xi)\cos 2\tau, \\ F_j(\tau) &= R_j + (1 - R_j)^2 \exp(-c\xi)[1 - R_j \exp(-c\xi)]^{-1}, \\ R_1 &= \left[\frac{\tan(\tau - \tau')}{\tan(\tau + \tau')}\right]^2, \qquad R_2 = \left[\frac{\sin(\tau - \tau')}{\sin(\tau + \tau')}\right]^2, \\ \xi &= (1 - n^{-2}\cos^2\tau)^{1/2},\end{aligned}$$

$\cos\tau' = \cos\tau/n$, $c = 2\alpha a$, $\alpha = 4\pi\chi/\lambda$ is the absorption coefficient of a particle substance. For simplicity we consider $\chi \ll n$. Equations (1)–(3) are much simpler than the solutions according to the Mie theory.

Nevertheless, Eqs. (1)–(3) are not convenient and simple enough for inverse-problem solutions, parameterization schemes in climate research, and so on.

Let us make a next step toward simplifying the problem. First, keep in mind that at $R_j \ll 1$ the function $F_j(\tau)$ [see Eq. (5)] can be represented in the form

$$F_j(\tau) = \exp(-c\xi) + R_j[1 - \exp(-c\xi)]^2, \tag{6}$$

where the terms $o(R_j{}^k)$ $(k \geq 2)$ are omitted. The contribution of the omitted terms to $F_j(\tau)$ is essential only over a small interval near $\tau \approx 0$, but the integrand in Eq. (5) is small in the vicinity of $\tau \approx 0$ because of the multiplier $\sin 2\tau$. Substituting Eq. (6) into Eq. (5) gives us

$$Q_{\mathrm{sca}}{}^{G} = \varphi(c) + S[1 - \exp(-bc)]^2, \tag{7}$$

where

$$\varphi(c) = 2n^2[(1 + cb)\exp(-cb) - (1 + c)\exp(-c)]c^{-2},$$

$$b = (1 - n^{-2})^{1/2}, \qquad S = \int_0^{\pi/2} R(\tau)\sin 2\tau \mathrm{d}\tau,$$

$$R(\tau) = (R_1 + R_2)/2. \tag{8}$$

The approximate equation

$$\frac{1}{2}\sum_{j=1}^{2}\int_0^{\pi/2} R_j[1 - \exp(-c\xi)]^2 \sin 2\tau \mathrm{d}\tau \approx [1 - \exp(-cb)]^2 S \tag{9}$$

was used to derive Eq. (7). Note that the parameters b and S depend on the index n alone. The value of S is the reflection coefficient from a plane interface with the refractive index n under diffuse illumination. Equations (1), (2), and (7) yield the following approximate solutions for the absorption cross section $C_{\mathrm{abs}} = C_{\mathrm{ext}} - C_{\mathrm{sca}}$:

$$C_{\mathrm{abs}}{}^{G} = \pi a^2\{1 - \varphi(c) - S[1 - \exp(-bc)]^2\}. \tag{10}$$

At $c \ll 1$, Eq. (10) gives us the well-known formula[6]

$$C_{\mathrm{abs}}{}^{G} = \alpha V\psi, \tag{11}$$

where $V = 4\pi a^3/3$ is the particle volume and $\psi = n^2(1 - b^3)$. At $c \gg 1$, from Eq. (10) we obtain[2]

$$C_{\mathrm{abs}}{}^{G} = \pi a^2(1 - S); \tag{12}$$

i.e., all the photons that penetrate into the particle are absorbed. As $n \rightarrow 1$, from Eq. (10) we obtain van de Hulst's formula[3]:

$$C_{\mathrm{abs}}{}^{G} = \pi a^2\{1 - 2c^{-2}[1 - (1 + c)\exp(-c)]\}. \tag{13}$$

So, in the particular cases of $c \rightarrow 0$, $c \rightarrow \infty$, and $n \rightarrow 1$, Eq. (10) reduces to known formulas[2,3,6] but also gives a solution for any intermediate values of $c = 2\alpha a$ and n.

Let us consider now the asymmetry parameter g^{G} of the phase function [see Eq. (3)] and represent it through two auxiliary values of g_0 and g_∞, which are the asymmetry parameters for nonabsorbing $(c = 0)$ and strongly absorbing $(c \rightarrow \infty)$ large spherical particles, respectively:

$$g^{G} = g_\infty - (g_\infty - g_0)\exp(-\eta c). \tag{14}$$

Analytical solutions for the functions S [see Eq. (8)] and g_∞ were obtained in Refs. 11 and 12. They are given in Appendix A. Approximations for the entire set of the auxiliary parameters S, g_∞, g_0, and η

obtained with the least-squares technique can be found in Ref. 13. Table 1 gives the values of these parameters for n ranging from 1.1 to 2.1. Let us emphasize that the additional error of Eqs. (7) and (14) in comparison with Eqs. (3) and (5) is no more than 1–2% for any value of the attenuation parameter c.

3. Correction for Edge Effects

A lot of natural aerosols (atmospheric aerosols, clouds in the infrared region of spectrum, and so on) contain particles with radii $a \approx 3\lambda$ to 5λ. Here the errors of the GO approximation are essential ($\geq 20\%$).[10,13] To diminish these errors the edge effects[3] should be taken into account. The edge terms that relied on an analytical transformation of Mie series were obtained in Ref. 10. Unfortunately, the results of Ref. 10 are too complicated for our purposes.

Let us choose another way and include the following differences:

$$C_{\text{ext}}{}^{E} = C_{\text{ext}} - C_{\text{ext}}{}^{G}, \tag{15}$$

$$C_{\text{abs}}{}^{E} = C_{\text{abs}} - C_{\text{abs}}{}^{G}, \tag{16}$$

$$g^{E} = g - g^{G}, \tag{17}$$

where the true values of C_{ext}, C_{abs}, and g are available through the Mie theory. By definition, the included functions $C_{\text{ext}}{}^{E}$, $C_{\text{abs}}{}^{E}$, and g^{E} give the contribution of the wave corrections (edge effects) to the values of the extinction and absorption cross sections and the asymmetry parameter. In view of the results from Refs. 3, 10, and 13, these correction terms could be represented as

$$C_{\text{ext}}{}^{E} = \frac{2\pi a^2}{x^{2/3}}, \quad C_{\text{abs}}{}^{E} = (n-1)[1-\exp(-1/\beta\rho)]C_{\text{abs}}{}^{G},$$
$$g^{E} = \frac{(\gamma_1 + \gamma_2 c)\exp(-\eta c)}{x^{2/3}}, \tag{18}$$

Table 1. Parameters S, g_0, g_∞, and η for Different n

n	S	g_0	g_∞	η
1.1	0.0252	0.9731	0.9946	0.5180
1.2	0.0443	0.9341	0.9856	0.6528
1.25	0.0529	0.9147	0.9806	0.6948
1.3	0.0611	0.8961	0.9751	0.7280
1.333	0.0664	0.8843	0.9714	0.7468
1.34	0.0675	0.8818	0.9706	0.7505
1.35	0.0691	0.8783	0.9695	0.7555
1.4	0.0768	0.8613	0.9638	0.7785
1.45	0.0844	0.8542	0.9579	0.7985
1.5	0.0918	0.8299	0.9520	0.8160
1.55	0.0991	0.8154	0.9460	0.8315
1.60	0.1063	0.8015	0.9400	0.8453
1.65	0.1133	0.7884	0.9340	0.8580
1.70	0.1203	0.7759	0.9280	0.8695
1.90	0.1475	0.7314	0.9046	0.9080
2.00	0.1606	0.7121	0.8933	0.9340
2.10	0.1734	0.6945	0.8823	0.9383

where $\rho = 2x(n-1)$ and β and γ_j are the unknown parameters that can depend both on n and on χ.

At $\rho \gg 1$ it follows from Eqs. (18) that

$$C_{\text{abs}}{}^{E} = \frac{1}{2\beta x} C_{\text{abs}}{}^{G}. \tag{19}$$

To find the coefficients β and γ_j in Eqs. (18), the computations for $C_{\text{abs}}{}^{E}$, g^{E}, and $C_{\text{ext}}{}^{E}$ were performed through Eqs. (15)–(17) by the use of the Mie theory and GO solutions [Eqs. (10) and (14)] at different n and χ. Comparison with the Mie calculations shows a high accuracy of the proposed Eqs. (18) for $x \geq 20$ and $\chi < 10^{-2}$, at least, for refractive indexes n ranging from 1.2 to 1.6 (the accuracy estimations are given in more detail below). Over this range the parameters β and γ_j could be taken independent of the n value [$\chi \in (10^{-2}, 10^{-5})$]

$$\beta = (21.2 + 20.1 \log \chi + 11.1 \log^2 \chi + \log^3 \chi)^{-1},$$
$$\gamma_1 = 0.5, \qquad \gamma_2 = 0.2. \tag{20}$$

So, the sought-for formulas are

$$C_{\text{ext}} = 2(1 + x^{-2/3})\pi a^2, \tag{21}$$

$$C_{\text{abs}} = T\left\{1 - \frac{2n^2}{c^2}[\exp(-cb)(1+cb) - \exp(-c)(1+c)] - S[1-\exp(-cb)]^2\right\}\pi a^2, \tag{22}$$

$$g = g_\infty - \left(g_\infty - g_0 + \frac{\gamma_1 + \gamma_2 c}{x^{2/3}}\right)\exp(-\eta c), \tag{23}$$

where $T = 1 + (n-1)[1 - \exp(-1/\beta\rho)]$.
The errors of Eqs. (21)–(23) are less than 15% at $x \geq 45$, $n = 1.2$–1.6, and $\chi < 10^{-4}$ [without one's averaging over the ripple oscillations of the functions $C_{\text{ext}}(x)$, $C_{\text{abs}}(x)$, and $g(x)$]. It is evident that averaging markedly decreases the computation error. This is shown below by the example of spherical polydispersions.

Let us estimate the contributions of the edge terms to the investigated optical parameters for different x, keeping in mind that our approach, in which the flux scattered by a particle is subdivided into parts because of diffraction, GO, and edge-effects components, has a physical meaning only at $a > \lambda$.

From Eq. (21) it follows that $q_1 = C_{\text{ext}}{}^{E}/C_{\text{ext}}{}^{G} = x^{-2/3}$, and the contribution of the edge term to the extinction cross section is approximately 10% at $x =$ 30%, 6% at $x = 60$. Let $n = 1.34$ and χ range between 10^{-4} and 10^{-2}. Then as the x value increases from 30 to 100, the ratio $q_2 = C_{\text{abs}}{}^{E}/C_{\text{abs}}{}^{G}$ changes from 30% to 8% as the ratio $q_3 = g^{E}/g^{a}$ changes from 6% to 0.1%. Naturally the contribution of the edge terms decreases with the increase of x. As can be seen from the intercomparison of q_i ($i = 1, 2, 3$) values, regard for the edge effects is most important for the absorption cross section C_{abs}.

4. Spherical Polydispersions

Solutions (21)–(23) provide an analytical generalization even for a polydispersed medium with some particle-size distribution (PSD) $f(a)$. For instance, let an aerosol be characterized by the gamma PSD[14,15]:

$$f(a) = Ba^{\mu} \exp(-\mu a/a_0), \qquad \int_0^{\infty} f(a)\mathrm{d}a = 1, \tag{24}$$

where $B = \mu^{\mu+1} a_0^{-(\mu+1)}/\Gamma(\mu + 1)$, Γ is the gamma function, a_0 is the mode radius, and μ is the half-width parameter of the PSD. In this case, in view of Eqs. (21) and (24) the extinction coefficient

$$\sigma_{\mathrm{ext}} = N \int_0^{\infty} C_{\mathrm{ext}}(a) f(a)\mathrm{d}a \tag{25}$$

is of the form

$$\sigma_{\mathrm{ext}} = \sigma_{\mathrm{ext}}{}^{G}\left[1 + \frac{D(\mu)}{x_{\mathrm{eff}}{}^{2/3}}\right]. \tag{26}$$

Here

$$\sigma_{\mathrm{ext}}{}^{G} = \frac{3C_v}{2a_{\mathrm{eff}}} \tag{27}$$

is the extinction coefficient within the GO approximation[2,3] and

$$a_{\mathrm{eff}} = \frac{\langle a^3 \rangle}{\langle a^2 \rangle}, \tag{28}$$

is the Sauter (or effective) radius of the PSD,[2,3]

$$\langle a^i \rangle = \int_0^{\infty} a^i f(a)\mathrm{d}a, \qquad x_{\mathrm{eff}} = k a_{\mathrm{eff}},$$

$$C_v = \frac{4\pi N}{3} \langle a^3 \rangle, \tag{29}$$

is the volumetric particle concentration, N is the number concentration, and

$$D(\mu) = (\mu + 3)^{2/3} \frac{\Gamma\left(\mu + \frac{7}{3}\right)}{\Gamma(\mu + 3)}. \tag{30}$$

D depends slightly on μ and $D(\mu) \rightarrow 1$ as $\mu \rightarrow \infty$ (see Table 2). To simplify Eq. (26), finally, one can take $D(\mu) = 1$, i.e.,

$$\sigma_{\mathrm{ext}} = \frac{1.5C_v}{a_{\mathrm{eff}}}\left[1 + \frac{1}{(k a_{\mathrm{eff}})^{2/3}}\right]. \tag{31}$$

Table 2. Function $D(\mu)$

μ	1	2	3	4	5	6	7	8	9	10
D	1.167	1.128	1.104	1.088	1.076	1.067	1.060	1.054	1.049	1.045

The relative error Δ of Eq. (31) in comparison with Eq. (26) is

$$\Delta = \frac{D(\mu) - 1}{D(\mu) + x_{\mathrm{eff}}{}^{2/3}}. \tag{32}$$

This relative error depends on μ and x_{eff}. As $x_{\mathrm{eff}} \rightarrow \infty$, $\Delta \rightarrow 0$ for any μ, and as $\mu \rightarrow \infty$, $\Delta \rightarrow 0$ for any x_{eff}. For example, at $x_{\mathrm{eff}} \geq 27$

$$\Delta \leq \frac{D(\mu) - 1}{D(\mu) + 9}, \tag{33}$$

e.g., $\Delta \leq 2\%$ at $\mu \geq 1$. So we can recommend simple Eq. (31) instead of Eq. (26).

Additionally, it should be noted that the extinction coefficient Eq. (31) depends on only two microphysical parameters, the volumetric concentration C_v and the effective radius a_{eff}. So Eq. (31) can be used for other types of PSD's without any changes (see below).

Let us now consider the absorption coefficient

$$\sigma_{\mathrm{abs}} = N \int_0^{\infty} C_{\mathrm{abs}}(a) f(a)\mathrm{d}a. \tag{34}$$

From Eqs. (22), (24), and (34) it follows that

$$\sigma_{\mathrm{abs}} = \sigma_{\mathrm{abs}}{}^{G} + \sigma_{\mathrm{abs}}{}^{E}, \tag{35}$$

where

$$\sigma_{\mathrm{abs}}{}^{G} = \frac{3C_v}{4a_{\mathrm{eff}}}[1 - \varphi_1(\bar{c}) - S\chi_1(\bar{c})] \tag{36}$$

is the absorption coefficient within the GO framework,

$$\varphi_1(\bar{c}) = 2n^2\bar{c}^{-2}[\nu_1(h_1{}^{\mu+1} - h_2{}^{\mu+1}) + \nu_2\bar{c}(bh_1{}^{\mu+2} - h_2{}^{\mu+2})], \tag{37}$$

$$\chi_1(\bar{c}) = h_4{}^{\mu+3} - 2h_1{}^{\mu+3} + h_3{}^{\mu+3}, \tag{38}$$

$$h_j = \left(1 + \frac{\bar{c}}{3 + \mu}\theta_j\right)^{-1}, \qquad j = 1, 2, 3, 4, \tag{39}$$

$$\theta_1 = b, \quad \theta_2 = 1, \quad \theta_3 = 2b, \quad \theta_4 = 0, \tag{40}$$

$$\nu_1 = \frac{\mu + 2}{\mu + 1}\nu_2{}^2, \quad \nu_2 = \frac{\mu + 3}{\mu + 2}, \quad \bar{c} = 2\alpha a_{\mathrm{eff}}, \tag{41}$$

and the integral

$$\sigma_{\mathrm{abs}}{}^{E} = N(n - 1)\int_0^{\infty} C_{\mathrm{abs}}{}^{G} \times \left[1 - \exp\left(-\frac{1}{2k(n - 1)\beta a}\right)\right] f(a)\mathrm{d}a \tag{42}$$

accounts for the edge effects. The analytical expression for the $\sigma_{\mathrm{abs}}{}^{E}$ value was obtained in Ref. 13 in terms of the gamma and the modified Bessel functions.

But it is too complicated so let us try to obtain a simpler solution for σ_{abs}. As was shown above for a given value of a_{eff}, the second term in Eq. (26) depends slightly on μ. Let us suppose that ${\sigma_{abs}}^E$ [see Eq. (42)] also mainly depends on a_{eff}, and for a given value of a_{eff} the μ dependence can be neglected. Then from Eq. (42) it follows approximately that

$$ {\sigma_{abs}}^E = {\sigma_{abs}}^G (n-1)\left\{1 - \exp\left[-\frac{1}{2k(n-1)\beta a_{eff}}\right]\right\}. \quad (43) $$

Finally, from Eqs. (35) and (43) it follows that

$$ \sigma_{abs} = \overline{T}{\sigma_{abs}}^G, \quad (44) $$

where

$$ \overline{T} = 1 + (n-1)[1 - \exp(-1/\beta\overline{\rho})], $$
$$ \overline{\rho} = 2ka_{eff}(n-1). \quad (45) $$

Let us consider the solution in Eq. (44) carefully. As $\overline{\rho} \to \infty$, it follows that $\sigma_{abs} \to {\sigma_{abs}}^G$, as it should be. As $\mu \to \infty$, one can find that ${h_1}^{\mu+1} \to {h_1}^{\mu+2} \to \exp(-\overline{c}b)$, ${h_2}^{\mu+1} \to {h_2}^{\mu+2} \to \exp(-\overline{c})$, $\chi_1 \to [1 - \exp(-\overline{c}b)]^2$, $\varphi_1(\overline{c}) \to \varphi(\overline{C})$, and, with regard to Eqs. (37) and (38), Eq. (35) reduces to Eq. (10). As $\overline{c} \to \infty$ (strong absorption), $h_j \to 0$ ($j = 1, 2, 3$), $\varphi_1(\overline{c}) \to 0$, and from Eq. (44) one can obtain

$$ \sigma_{abs} = \frac{3C_v\overline{T}}{4a_{eff}}(1 - S). \quad (46) $$

As $\overline{c} \to 0$ (weak absorption), it follows from Eq. (44) that

$$ \sigma_{abs} = \alpha\psi(n)\overline{T}C_v[1 + \alpha a_{eff}\overline{\psi}(n)], \quad (47) $$

where

$$ \overline{\psi}(n) = 3(2n^2 - 1)/[4n^4(1 - b^3)]. $$

Equation (47) coincides with the known Bohren equation[6] at $\rho \to \infty$ ($\overline{T} = 1$) and $\alpha a_{eff} = 0$. But additionally Eq. (47) provides a higher accuracy in the computation of the absorption coefficient of spherical polydispersions composed of intermediate-size particles. Moreover, the solution that is found takes into account the quadratic term of series expansion with respect to the parameter α. Note again that both Eqs. (46) and (47) depend on only two microphysical parameters, a_{eff} and C_v [see also Eq. (31)].

In conclusion, let us derive a simple analytical formula for the asymmetry parameter $\langle\cos\theta\rangle$ of a spherical polydispersion:

$$ \langle\cos\theta\rangle = \frac{\int_0^\infty g(a)C_{sca}(a)f(a)\mathrm{d}(a)}{\int_0^\infty C_{sca}(a)f(a)\mathrm{d}(a)}. \quad (48) $$

In view of Eqs. (21)–(24), from Eq. (47) it follows approximately that

$$ \langle\cos\theta\rangle = \langle\cos\theta\rangle^G - \langle\cos\theta\rangle^E, \quad (49) $$

where

$$ \langle\cos\theta\rangle^G = g_\infty - (g_\infty - g_0)\frac{h_2 + \varphi_2 + x_2 S}{1 + \varphi_1 + x_1 S} \quad (50) $$

is the GO value of the asymmetry parameter. Equations for the functions φ_2 and χ_2 can be obtained from Eqs. (37) and (38), respectively, for the functions φ_1 and χ_1, replacing the values of θ_j [see Eq. (40)] in these equations by the values of $(\theta_j + \eta)$. The function $\langle\cos\theta\rangle^E$ accounts for the edge effects. As above, let us suppose that $\langle\cos\theta\rangle^E$ depends slightly on μ for a given value of a_{eff}. With this in mind, one can obtain [see Eq. (18)]

$$ \langle\cos\theta\rangle^E = \frac{\gamma_1 + \gamma_2\overline{c}}{{x_{eff}}^{2/3}}\exp(-\eta\overline{c}). \quad (51) $$

To obtain Eq. (49) from Eq. (48), we have ignored the small terms, which are the products of the edge terms. A more accurate (but complex) formula for $\langle\cos\theta\rangle$ was derived in Ref. 13 through the use of some special functions.

It should be emphasized that, as $\overline{c} \to \infty$ and as $\overline{c} \to 0$, more simple solutions follow from Eqs. (49)–(51). From Eq. (49) one can obtain $\langle\cos\theta\rangle = g_\infty$ [see Eq. (14)] for a large $\overline{c}$ value and

$$ \langle\cos\theta\rangle = g_0 - \frac{\gamma_1 + \gamma_2\overline{c}}{{x_{eff}}^{2/3}} + \eta\overline{c}\left(g_\infty - g_0 + \frac{\gamma_1}{{x_{eff}}^{2/3}}\right) \quad (52) $$

for a small $\overline{c}$ value.

5. Accuracy of Approximations

Comparison between the data obtained through Eqs. (26), (35), and (49), and the Mie computations was made at $\lambda = 0.55$ µm; $n = 1.2, 1.34, 1.55$; $\chi = 0.10^{-4}, 10^{-3}, 10^{-2}$; $a_0 = 2.5$ and 5 µm; and $\mu = 2, 6, 8$. In so doing, the algorithm[16] was used to calculate the Mie series. It was found that the relative error Δ of the extinction-coefficient estimation fluctuates near zero and does not exceed 1% over the entire considered range. This is the reason why the error of the value of $\delta = 1 - \omega_0$ (where $\omega_0 = \sigma_{sca}/\sigma_{ext}$ is the single-scattering albedo) is mainly defined by the error of the absorption-coefficient computation. As for the parameters $1 - \langle\cos\theta\rangle$ and σ_{abs}, the errors of their computation are less than 10% over the entire studied range. At $\chi = 10^{-4}$, $n = 1.34$, $a_0 = 2.5$ µ, and $\mu = 6$, the relative error of the δ estimate with regard to the edge effects is less than 2%, although without such a condition δ is approximately 16%.

The contribution of the edge terms becomes more and more essential when the wavelength increases. For instance, in the near-infrared region of spectrum the extinction coefficient of the clouds increases when

the wavelength grows. This feature was noted by Deirmendjian.[14] As is known, the extinction coefficient within the framework of the GO approximation shows no spectral dependence. Unlike the GO approximation, an accounting of the edge terms in our solutions gives the correct spectral dependence of the extinction coefficient.

It should be emphasized that it is no mere chance that the accuracy of the values of $(1 - \omega_0)$ and $(1 - \langle\cos\theta\rangle)$ was discussed above. As a matter of fact, the accuracy of the estimations of the radiative characteristics of weakly absorbing layers depends on the accuracy of the determination of just these small values.[17]

The convenient analytical formulas for the extinction and absorption coefficients of clouds were obtained in Refs. 17 and 18 with the van de Hulst anomalous diffraction approximation, with allowance for the refraction of light by drops. The relative errors of the single-scattering albedo and the asymmetry parameters $\langle\cos\theta\rangle$ of the water-cloud and dust particles were found to be less than 3–5%. In this case the errors of $(1 - \omega_0)$ and $(1 - \langle\cos\theta\rangle)$ that are responsible for the radiation characteristics (reflection and transmission functions, radiative fluxes, etc.) are essentially higher (up to $\pm 30\%$). Apparently, for atmospheric aerosols in the visible region of the spectrum for which the refractive index ranges from 1.3 to 1.7,[14] it is just impossible to provide a high-accuracy test of the estimation of the optical parameters on the basis of the anomalous diffraction theory.

So we believe that our approximation (which provides the values of $1 - \omega_0$ and $1 - \langle\cos\theta\rangle$ with errors of no more than 8–10%) is good enough for radiation computations.

Although they were accurate enough and much simpler than Mie formulas, Eqs. (26), (35), and (49) are still rather cumbersome. But these solutions can yield desired simple expressions for the optical parameters in many cases.

As mentioned above, one of the most important fields in which the obtained formulas can be applied is the problem of parameterization of cloud–radiation interaction. Let us consider this case more carefully.

As is known,[15] the effective radius α_{eff} of droplets of continental clouds usually ranges from 3 to 12 μm, and the half-width parameter μ ranges from 2 to 8. For $\lambda \le 2.25$ μm, the imaginary part of the refractive index of water is rather small ($\chi x_{\mathrm{eff}} \to 0$), and Eqs. (47) and (52) can be used.

The values of g_0, g_∞, and η [see Eq. (52)] are given in Table 1. Note that the real part of the refractive index of water in the near-infrared region of the spectrum changes slowly ($n = 1.29$–1.34). Within this range of n, the values of η, g_∞, and g_0 change slowly, too. They can be replaced by constants: we have used the values $\eta(1.34)$, $g_0(1.34)$, and $g_\infty(1.34)$ (see Table 1) for all λ in the near-infrared and the visible regions of the spectrum. With this in mind, from Eq. (52) it follows that

$$1 - \langle\cos\theta\rangle = 0.1182 + \frac{0.5 + 0.2\bar{c}}{{x_{\mathrm{eff}}}^{2/3}} - 0.75\bar{c}\left(0.09 + \frac{1}{2{x_{\mathrm{eff}}}^{2/3}}\right). \qquad (53)$$

Equations (26), (47), and (53) [$D(\mu) = 1.1$ at $\mu = 2$–8 (see Table 2)] can be used for computations of the local optical cloud properties in the visible and the near-infrared regions of the spectrum ($\lambda \le 2.25$ μm).

Note that all the optical parameters of clouds that have been considered depend on only one PSD parameter, namely, a_{eff}. This interesting feature of cloud optics has been mentioned more than once,[19] but the obtained solution presents it clearly. Comparison between the data obtained through Eqs. (26), (47), and (53) at $D(\mu) = 1.1$ and through the Mie theory was performed for water droplets at $\lambda = 0.45$–2.25 μm for two types of PSD: the gamma PSD [see Eq. (24)] at $a_0 = 3, 4, 8$ μm; $\mu = 2, 6$; and the log-normal PSD

$$f(a) = \exp[-\ln^2(a/a^*)/2\sigma^2]/\sqrt{2\pi}a\sigma \qquad (54)$$

at $a^* = 4.49$ μm and $\sigma = 0.34$ ($a_{\mathrm{eff}} = 6$ μm). The relative errors of the approximations for the values of σ_{ext}, σ_{abs}, $1 - \omega_0$, and $1 - \langle\cos\theta\rangle$ in all cases are less than 5–8%.

6. Conclusion

We propose analytical formulas for the asymmetry parameters and the extinction and absorption coefficients of spherical polydispersions that include diffraction, geometric-optics terms, and edge terms. These formulas provide a sufficiently high accuracy.

The especially simple approximations for the optical parameters of clouds were derived. The values of $\sigma_{\mathrm{ext}}/C_v$, $1 - \omega_0$ and $1 - \langle\cos\theta\rangle$ depend on only one microstructure parameter, a_{eff}, and agree with the Mie computation to within 5–8% for typical water-cloud models in the near-infrared and the visible regions of the spectrum up to $\lambda = 2.25$ μm.

The proposed formulas appear to be very useful in different applications, including the direct and inverse problems of cloud and aerosol optics.

Appendix A.

To present a complete picture, in this section the analytical expressions for the auxiliary functions [see Eqs. (22) and (23)] found in Refs. 11 and 12 at $\chi \ll n$ are given:

$$S = \int_0^{\pi/2} R(\tau)\sin 2\tau \mathrm{d}\tau, \qquad (A1)$$

$$g_\infty = \frac{1 + \frac{1}{2}\int_0^{\pi/2} R(\tau)\sin 4\tau \mathrm{d}\tau}{1 + S}. \qquad (A2)$$

These functions may be rewritten in a more instruc-

tive form. So for the function S we have

$$S = S_1 \ln n - S_2 \ln \frac{n+1}{n-1} + S_3, \tag{A3}$$

where

$$S_1 = \frac{8n^4(n^4+1)}{(n^4-1)^2(n^2+1)}, \qquad S_2 = \frac{n^2(n^2-1)^2}{(n^2+1)^3},$$

$$S_3 = \frac{\sum_{j=0}^{7} A_j n^j}{3(n^4-1)(n^2+1)(n+1)},$$

$$A_j = (-1, -1, -3, 7, -9, -13, -7, 3).$$

A solution for g_∞ is of the form

$$g_\infty = \frac{1 + g_1 \ln n - g_2 \ln[(n+1)/(n-1)] + g_3}{1+S}, \tag{A4}$$

where

$$g_1 = \frac{8n^4(n^6 - 3n^4 + n^2 - 1)}{(n^4-1)^2(n^2+1)^2},$$

$$g_2 = \frac{(n^2-1)^2(n^8 + 12n^6 + 54n^4 - 4n^2 + 1)}{16(n^2+1)^4},$$

$$g_3 = \frac{\sum_{j=1}^{12} B_j n^j}{24(n^2+1)^2(n^4-1)(n+1)},$$

$$B_j = (-3, 13, -89, 151, 186, 138, -282, 22, 25, 25, 3, 3).$$

Equations (A3) and (A4) can be used to avoid the numerical computation of the corresponding integrals in governing Eqs. (22), (23), (35), and (49).

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